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**BRL****PHYSICAL BASIS FOR A PRESSURE-DEPENDENT  
YIELD CRITERION FOR POLYMERS**

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LAWRENCE LIVERMORE NATIONAL LABORATORY

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## 1. INTRODUCTION

Structural applications for fiber-reinforced polymer composites demand that the thermoplastic or thermoset matrix materials survive complex multiaxial loading and severe environmental effects without causing failure of the composite part. Consequently, it is important to determine a failure or yield criterion for the constituent polymer matrix materials under a generalized stress state. This information in conjunction with modeling efforts on the state of stress in fiber-reinforced composites should provide insight into the poorly understood relationship between matrix- and composite-failure. Along these lines, the present work is an attempt to obtain a more basic understanding of multiaxial yielding in homogeneous polymeric solids.

In every relevant study to date, polymeric solids have been shown to exhibit significant increases in yield stress with pressure. For this reason, a yield criterion for polymers must include the effect of pressure or mean normal stress. One criterion which has the correct form to describe the observed variation of yield stress with pressure for polymeric solids is

$$J_2' = \sum_{i=0}^N \alpha_i J_1^i \quad (1)$$

which was originally proposed by Hu and Pae (1963). In Equation 1,  $J_2'$  is the second invariant of stress deviators,  $J_2' = 1/2 \sigma_{ij}' \sigma_{ij}'$  ( $i, j = 1, 2, 3$ ), and  $J_1$  the first stress invariant given by  $J_1 = \sigma_{kk} = \sigma_{11} + \sigma_{22} + \sigma_{33}$ . The deviatoric stresses are  $\sigma_{ij}' = \sigma_{ij} + p\delta_{ij}$ , for a mean normal pressure  $p = -1/3 \sigma_{kk}$ .

For  $N = 0$ , Equation 1 results in the distortional energy criterion proposed by Huber (1904) and Hencky (1924)

$$J_2' = \alpha_0 = k^2, \quad (2)$$

where the constant,  $k$ , is the yield stress determined in simple shear. Equation 2 has a physical basis in the assumption that a critical value of the distortional part of the strain energy determines the onset of plastic deformation (i.e., uniform pressure has no effect on yielding). Eichinger (1926) showed that the distortional energy criterion (Equation 2) is equivalent to the statement that yielding occurs when the shear stress on the octahedral plane,  $\tau_{oct}$ , reaches the critical value,  $\tau_{oct} \equiv (2J_2'/3)^{1/2} = 0.816 k$ . Von Mises (1913) assumed that a critical value of the mean difference in the principle stresses caused yielding and proposed that  $(J_2')^{1/2} = k$ . Although derived from different assumptions, each of the distortional energy, octahedral

shear stress, and von Mises criteria predict the same yield behavior and find great utility in describing the onset of plastic deformation in ductile metals at moderate pressures.

In contrast to metals, polymeric solids generally exhibit highly pressure-dependent yield behavior as evidenced by a significant increase in yield stress with hydrostatic pressure and a lower yield stress in simple tension than in simple compression. One way to account for these pressure effects is to expand  $J'_2$  as a polynomial in  $J_1$ , i.e.,

$$J'_2 = k^2 + \alpha_1 J_1 \quad (3)$$

or

$$J'_2 = k^2 + \alpha_1 J_1 + \alpha_2 J_1^2, \quad (4)$$

which correspond to Equation 1 with  $N = 1$  and  $N = 2$ , respectively. By analogy with Equations 2, Equations 3 and 4 are modified distortional energy yield criteria and have been used successfully to describe pressure-dependent yielding for a number of polymers (Mears, Pae, and Sauer 1969; Sauer, Mears, and Pae 1970).

Equation 3, in slightly different form (Raghava, Caddell, and Gregory 1973), was compared by Caddell, Raghava, and Adkins (1974) and Matsushige, Radcliffe, and Baer (1975) to a modified octahedral shear stress yield criterion for polymers proposed by Sternstein and Ongchin (1969), Bauwens (1970), and with minor variation by Spitzig and Richmond (1979). It was found that to first order in pressure, the inherently nonlinear, modified distortional energy criterion provided a generally better fit to the yield stress data for all of the polymers examined than did the modified octahedral shear stress/von Mises criterion—which predicts a linear variation of yield stress with pressure according to  $(J'_2)^{1/2} = k + aJ_1$ . To obtain better agreement with high pressure data, Silano, Bhateja, and Pae (1979) expanded the von Mises criterion as a power series in  $J_1$  (i.e.,  $[J'_2]^{1/2} = k + aJ_1 + bJ_1^2 + \dots$ ), achieving comparable predictive capability to the modified distortional energy criterion (Silano, Bhateja, and Pae 1974; Silano, Pae, and Sauer 1977; Pae 1977).

In contrast to the modified von Mises criterion, the modified distortional energy criterion maintains the dimensionality of a critical energy theory. Haigh (1970) proposed a total strain energy criterion for failure which includes both distortional and dilatational mechanical energy. In the above terminology, the total strain energy criterion takes the form:  $J'_2 = k^2 + \alpha_2 J_1^2$ , which unlike Equation 3 or 4 does not contain

the first-order pressure term,  $\alpha_i J_1$ , necessary to describe the observed variation of polymer yield stress with pressure. Consequently, within the context of the infinitesimal, equilibrium theory, a mechanical energy criterion cannot account for polymer yielding under a generalized state of stress. Recognizing that mechanical energy is only one form of free energy, the following sections examine the possibility that a different thermodynamic potential—the internal energy—may be the criterion for polymer yielding.

## 2. THERMODYNAMIC YIELD CRITERION

For a reversible solid deformation process, the combined first and second laws of thermodynamics can be written for a closed system at uniform temperature,  $T$ ,

$$dU = TdS + \sigma_{ij} d\epsilon_{ij} , \quad (5)$$

where  $dU$ ,  $dS$  are incremental changes in the internal energy and entropy per unit volume, respectively, and  $\epsilon_{ij}$  are the components of the small strain tensor

$$\epsilon_{ij} = \frac{1}{2} \left[ \frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right] . \quad (6)$$

Following Sneddon (1974), we consider that  $U = U(\epsilon_{ij}, T)$ , so that the total differential for internal energy is written

$$dU = \left( \frac{\partial U}{\partial \epsilon_{ij}} \right)_T d\epsilon_{ij} + \left( \frac{\partial U}{\partial T} \right)_{\epsilon_{ij}} dT . \quad (7)$$

The Helmholtz free energy,  $A$ , is defined

$$A = U - TS . \quad (8)$$

Writing Equation 8 in incremental form and substituting Equation 5,

$$dA = \sigma_{ij} d\epsilon_{ij} - SdT , \quad (9)$$

from which the following relationships are obtained:

$$\left( \frac{\partial A}{\partial \epsilon_{ij}} \right)_T = \sigma_{ij}; \quad \left( \frac{\partial A}{\partial T} \right)_{\epsilon_{ij}} = -S, \quad (10)$$

showing that

$$\left( \frac{\partial \sigma_{ij}}{\partial T} \right)_{\epsilon_{pq}} = - \left( \frac{\partial S}{\partial \epsilon_{ij}} \right)_T. \quad (11)$$

From Equations 5 and 11,

$$\left( \frac{\partial U}{\partial \epsilon_{ij}} \right)_T = \sigma_{ij} - T \left( \frac{\partial \sigma_{ij}}{\partial T} \right)_{\epsilon_{pq}}. \quad (12)$$

In addition, it can be shown that

$$\left( \frac{\partial U}{\partial T} \right)_{\epsilon_{ij}} = T \left( \frac{\partial S}{\partial T} \right)_{\epsilon_{ij}} = C_e, \quad (13)$$

where  $C_e$  is the heat capacity of the solid at constant strain. Substituting Equations 12 and 13 into Equation 7 gives the variation in internal energy density for incremental changes in strain and temperature as

$$dU = \left[ \sigma_{ij} - T \left( \frac{\partial \sigma_{ij}}{\partial T} \right)_{\epsilon_{pq}} \right] d\epsilon_{ij} + C_e dT, \quad (14)$$

which provides the necessary relationship to calculate the internal energy change for a solid deformation if the constitutive equation for the material is known.

The constitutive relationship for an isotropic, linear thermoelastic solid can be written

$$\sigma_{ij} = \frac{E}{(1+v)(1-2v)} [v e \delta_{ij} + (1-2v) \epsilon_{ij} - (1+v) \beta (T - T_o) \delta_{ij}], \quad (15)$$

with  $E$  and  $\beta$  the Young's modulus and linear thermal expansion coefficient of the material having Poisson's ratio,  $\nu$ . The dilatation,  $e$ , is  $e = \delta V/V_o = \epsilon_{ii}$ . Substitution of Equation 15 into Equation 14 and integrating gives the internal energy for a linear thermoelastic solid, in general, as

$$U - U_o = \frac{1}{2} \sigma_{ij} \epsilon_{ij} + \frac{E\beta(T + T_o)e}{2(1 - 2\nu)} + C_e(T - T_o) . \quad (16)$$

where  $U_o$  denotes the internal energy per unit volume of the reference state,  $\epsilon_{ij} = 0$ ,  $T = T_o$ . The second term on the right-hand side of Equation 16 results from the coupling of elastic deformation and thermal diffusion, while the third term represents the heat content. In contrast to the free energy, the internal energy includes thermoelastic coupling even at the reference temperature,  $T = T_o$ .

The first term on the right-hand side of Equation 16 is the strain energy density which can be decomposed into distortional and dilatational components via

$$\frac{1}{2} \sigma_{ij} \epsilon_{ij} = G \epsilon'_{ij} \epsilon'_{ij} + \frac{1}{2} K \epsilon_{ii} \epsilon_{kk} , \quad (17)$$

with  $G$  and  $K$  the shear and bulk modulus, respectively. The deviatoric strain is  $\epsilon'_{ij}$ , which is defined  $\epsilon'_{ij} = \epsilon_{ij} - e/3 = \epsilon_{ij} - \epsilon_{ii} \delta_{ij}/3$ . Since  $\sigma'_{ij} = 2G\epsilon'_{ij}$ , and  $\sigma_{kk} = 3K\epsilon_{ii}$ , Equation 17 can be written as

$$\frac{1}{2} \sigma_{ij} \epsilon_{ij} = \frac{1}{4G} \sigma'_{ij} \sigma'_{ij} + \frac{\sigma_{kk}^2}{18K} . \quad (18)$$

Thermoelastic coupling is reflected in the second term on the right-hand side of Equation 16 which can be expressed as

$$\frac{E\beta(T + T_o)e}{2(1 - 2\nu)} = \frac{1}{2} \beta(T + T_o) \sigma_{kk} . \quad (19)$$

Substituting Equations 18 and 19 into Equation 16 results in an expression for the internal energy change for an isothermal deformation in terms of the stress components

$$U - U_o = \frac{1}{4G} \sigma'_{ij} \sigma'_{ij} + \frac{1}{2} \beta(T + T_o) \sigma_{kk} + \frac{\sigma_{kk}^2}{18K} + C_e(T - T_o) . \quad (20)$$

In terms of the stress invariants, the internal energy change takes the form

$$U - U_o = \frac{1}{2G} J_2' + \frac{1}{2} \beta (T + T_o) J_1 + \frac{1}{18K} J_1^2 + C_e (T - T_o) . \quad (21)$$

The assumption is now made that the criterion for yielding of an isotropic polymeric solid in a state of combined stress is that the mechanical part of the internal energy density of the material reaches some critical value. In other words, we neglect the heat content and of the material and consider only that part of the internal energy which involves mechanical coupling with the outside world. Partitioning the internal energy according to  $U - U_o = \Delta + C_e (T - T_o)$ , the mechanical part of the internal energy,  $\Delta$ , is obtained from Equation 21 as

$$\Delta = \frac{1}{2G} J_2' + \frac{1}{2} \beta (T + T_o) J_1 + \frac{1}{18K} J_1^2 . \quad (22)$$

Considering only the mechanical internal energy as contributing to failure eliminates the unlikely possibility that a pure heating process can cause yielding in the absence of stress.

The critical value of the mechanical internal energy at yield can be determined from a simple shear experiment where  $J_1 = 0$ , so that  $J_2' = \sigma_{12}^2 = \tau_y^2$  at the onset of plastic deformation, and Equation 22 becomes

$$\Delta = \tau_y^2/2G = k^2/2G . \quad (23)$$

The condition for isothermal yielding at temperature,  $T$ , in a general stress state is finally obtained from Equations 22 and 23 as

$$J_2' = k^2 - [G\beta(T + T_o)]J_1 - [G/9K]J_1^2 , \quad (24)$$

which is the empirical Equation 4, with the pressure coefficients now defined as  $\alpha_1 = -G\beta(T + T_o)$  and  $\alpha_2 = -G/9K$ .

The implication of a critical value of the mechanical internal energy density at yield can be demonstrated graphically for a uniaxial deformation. If the coordinate axes are chosen such that the

stresses are in the principle directions, a uniaxial tension or compression in the 1 direction has  $\sigma_{11} = \pm\sigma_1$ ,  $\sigma_{22} = \sigma_{33} = 0$ , so that Equation 22 can be written

$$\Delta = \frac{1}{2} \beta (T + T_o) \sigma_1 + \frac{1}{2E} \sigma_1^2 . \quad (25)$$

The quadratic relationship between mechanical internal energy and uniaxial stress for an isothermal deformation is shown schematically in Figure 1. Stipulating a critical value for the mechanical internal energy at yield requires that the magnitude of the yield stress in simple compression be larger than the yield stress in simple tension. This mean normal pressure effect on uniaxial yielding is described by Equation 24.

The predicted negative sign of the  $J_1$  coefficient,  $\alpha_1$  in Equation 24 is in agreement with every relevant experimental study of polymer yielding. The magnitude of  $\alpha_1$  for engineering thermoplastics at  $T = T_o = 295^\circ K$  can be calculated using literature values for  $\beta$  and  $G$  (van Krevelen 1976) to be of the order  $\alpha_1 = -2G\beta T_o \approx -25 \pm 10 \text{ MPa}$ , which is roughly 10 times larger than typical experimental values of  $\alpha_1 \approx -3 \pm 2 \text{ MPa}$  (Mears, Pae, and Sauer 1969; Sauer, Mears, and Pae 1970; Raghava, Caddell, and Gregory 1973; Caddell, Raghava, and Adkins 1974; Matsushige, Radcliffe, and Baer 1975; Sternstein and Ongchin 1969; Bauwens 1970; Spitzig and Richmond 1979; Silano, Bhateja, and Pae 1974; Silano, Pae, and Sauer 1977; Pae 1977) obtained for various thermoplastics. The magnitude of the  $J_1^2$  coefficient,  $\alpha_2$ , is estimated to be small and on the order of  $\alpha_2 = -G/9K \approx -0.02$ , which compares favorably with the measured  $\alpha_2 = -0.010$  and  $-0.017$  for polyethylene and polypropylene, respectively (Mears, Pae, and Sauer 1969). However, values (Sauer, Mears, and Pae 1970) for polytetrafluoroethylene ( $\alpha_2 = 1.2 \times 10^{-5}$ ) and polycarbonate ( $\alpha_2 = 5.1 \times 10^{-4}$ ) are positive and significantly smaller than predicted by Equation 24.

### 3. DISCUSSION

The physical basis for a pressure-dependent yield criterion for polymers may be a critical value of the mechanical part of the internal energy density at the onset of plastic deformation. With this assumption, an isothermal yield condition is obtained from linear thermoelasticity and equilibrium thermodynamics in terms of the stress invariants, temperature, and material properties which has the same form as an experimentally proven polynomial expansion to second order in pressure.

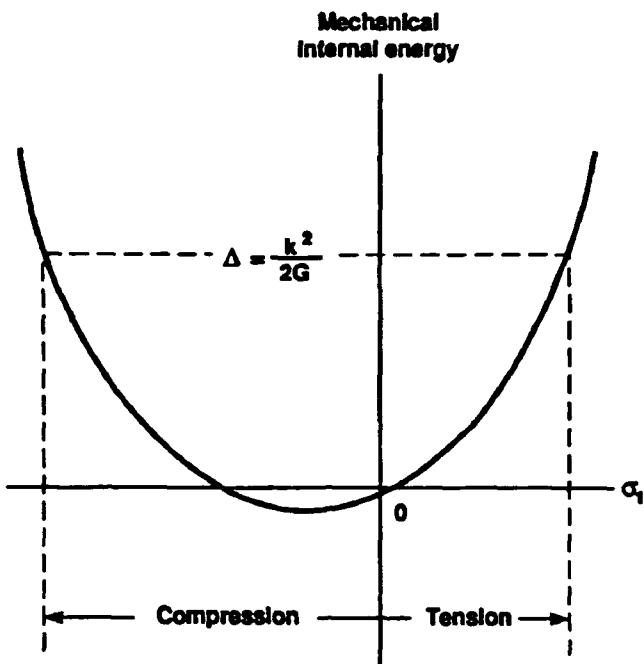


Figure 1. Schematic Showing Relationship Between Mechanical Internal Energy and Uniaxial Stress for an Isothermal Deformation.

Order of magnitude agreement between predicted and published experimental values of the pressure coefficients is observed. This result is encouraging, considering the highly nonlinear, nonequilibrium behavior of typical engineering thermoplastics prior to yielding and the characteristically large strains ( $\epsilon = 4$  to 5%) at which fully plastic deformation commences. Moreover, the linear theory does not address the universally observed pressure dependence of the shear, tensile (Sauer 1977; Pae and Bhateja 1975), and bulk (Bhateja and Pae 1975) moduli, nor the possibility of premature failure due to inhomogeneous, localized deformation (Sternstein, Ongchin, and Silverman 1968).

It can be shown (Lyon and Farris 1987) that, for an isothermal deformation at the reference temperature, the linear term in  $J_1$  appearing in Equation 24 corresponds to the reversible heat of deformation (i.e., the entropic component of the internal energy). Considering the statistical nature of macromolecules, it is reasonable that a successful yield criterion for polymer solids should reflect the entropic origin of chain deformation.

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